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## High $^3\text{He}/^4\text{He}$ ratios in picritic basalts from Baffin Island and the role of a mixed reservoir in mantle plumes

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The high  $^3\text{He}/^4\text{He}$  ratio of volcanic rocks thought to be derived from mantle plumes is taken as evidence for the existence of a mantle reservoir that has remained largely undegassed since the Earth's accretion<sup>1–3</sup>. The helium isotope composition of this reservoir places constraints on the origin of volatiles within the Earth and on the evolution and structure of the Earth's mantle. Here we show that olivine phenocrysts in picritic basalts presumably derived from the proto-Iceland plume at Baffin Island, Canada, have the highest magmatic  $^3\text{He}/^4\text{He}$  ratios yet recorded. A strong correlation between  $^3\text{He}/^4\text{He}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $^{143}\text{Nd}/^{144}\text{Nd}$  and trace element ratios demonstrate that the  $^3\text{He}$ -rich end-member is present in basalts that are derived from large-volume melts of depleted upper-mantle rocks. This reservoir is consistent with the recharging of depleted upper-mantle rocks by small volumes of primordial volatile-rich lower-mantle material at a thermal boundary layer between convectively isolated reservoirs. The highest  $^3\text{He}/^4\text{He}$  basalts from Hawaii and Iceland plot on the observed mixing trend. This indicates that a  $^3\text{He}$ -recharged depleted mantle (HRDM) reservoir may be the principal source

of high  $^3\text{He}/^4\text{He}$  in mantle plumes, and may explain why the helium concentration of the 'plume' component in ocean island basalts is lower than that predicted for a two-layer, steady-state model of mantle structure.

Ocean island basalts (OIB) derived from mantle plumes commonly have  $^3\text{He}/^4\text{He}$  ratios that are higher than mid-ocean ridge basalts (MORB) that originate in the upper, asthenospheric mantle<sup>1–3</sup>. High  $^3\text{He}/^4\text{He}$  and solar-like Ne isotope ratios<sup>4</sup> reflect a higher proportion of primordial volatiles in the source region and, in the current paradigm, indicate that plumes tap a deep mantle reservoir that is significantly less degassed than the asthenospheric mantle. Although a number of locations have been proposed for this reservoir<sup>5</sup>, a lower mantle that has been convectively isolated below the 670-km seismic discontinuity for the lifetime of the Earth<sup>6</sup> is consistent with the mass balance of the depleted mantle and continental crust<sup>7</sup>. However, layered mantle models are seemingly at odds with seismic studies that demonstrate convective flow across the 670-km boundary<sup>8</sup> and require a 100- to 200-fold depletion of the He concentration in the upper, degassed mantle relative to the deep undegassed mantle reservoir<sup>6</sup>. Plume-derived OIB from, for example, Loihi seamount, Hawaii, have lower He concentrations than basalts from the degassed upper mantle<sup>2</sup> and have posed a persistent problem for a unified geochemical model of Earth structure<sup>6</sup>. The apparent paradox can be explained, at least in part, by more extensive degassing of ocean island basalts<sup>9</sup>. However, basalts from Iceland and the Hawaiian islands have linear, rather than strongly hyperbolic, He–Pb isotope mixing arrays that are consistent with relatively small He concentration contrasts between the degassed mantle and less degassed (high  $^3\text{He}/^4\text{He}$ ) mantle reservoirs<sup>9,10</sup>.

Picritic basalts erupted on Baffin Island, northeast Canada, 61 Myr ago correlate with the Anaanaa Member of the Vaigat Formation in West Greenland<sup>11</sup>, and are among the earliest manifestations of the ancestral Iceland mantle plume<sup>12</sup>. Trace-element and Sr–Nd isotope ratios show that they were generated by mixing of two discrete sources; relatively depleted mantle with  $(\text{La}/\text{Sm})_n < 1$ ,  $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7030$ ,  $^{143}\text{Nd}/^{144}\text{Nd} \approx 0.5130$ , and relatively enriched mantle with  $(\text{La}/\text{Sm})_n > 1$ ,  $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.7042$  and  $^{143}\text{Nd}/^{144}\text{Nd} \approx 0.51282$  (ref. 13). The depleted basalts are similar to the most depleted Icelandic basalts and North Atlantic MORB<sup>14</sup>. The enriched basalts are similar to, though slightly more extreme (higher  $^{87}\text{Sr}/^{86}\text{Sr}$  and lower  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios) than the most enriched basalts erupted by the Iceland plume and across the North Atlantic igneous province<sup>15</sup>.

The isotopic composition of He released by *in vacuo* crushing olivine phenocrysts from picritic basalts from three stratigraphically distinct cliff sections in northeast Baffin Island (Table 1) ranges from  $14.4R_a$  to  $49.5R_a$  (where  $R_a$  is the atmospheric  $^3\text{He}/^4\text{He}$  ratio;  $1.39 \times 10^{-6}$ ). These ratios are far higher than MORB values and extend the range measured in contemporaneous picrites from West Greenland<sup>16</sup>. The high ratios cannot result from contamination by *in situ* cosmogenic  $^3\text{He}$  because (1) *in vacuo* crushing does not release cosmogenic He from the olivine lattice<sup>17</sup>, (2) cosmogenic  $^3\text{He}$  production was negligible because the samples were collected from eroding, near-vertical sea cliff faces, and (3) the  $^3\text{He}/^4\text{He}$  of powders produced by crushing are systematically lower than crush values (Table 1) indicating that radiogenic He dominates the olivine lattice. Nucleogenic  $^3\text{He}$  production from  $^6\text{Li}$  is negligible in Palaeocene basalts<sup>16</sup>.

Crush release of radiogenic  $^4\text{He}$  in the olivine lattice and melt inclusions by post-eruptive U and Th decay tends to lower  $^3\text{He}/^4\text{He}$ , so measured ratios may underestimate the source  $^3\text{He}/^4\text{He}$ . Any such effect would be most pronounced in samples with the lowest He content.  $^3\text{He}/^4\text{He}$  are not correlated with He concentration (Table 1), implying that post-eruptive radiogenic He is insignificant to the crush-release He. Crustal contamination of the basalts by amphibolite- and granulites-facies basement rocks would also lower

Table 1 He, Sr and Nd isotope ratios, and trace-element data for Baffin Island basalts

Sample	$^4\text{He}^*$ ( $10^{-9}\text{cm}^3\text{STP g}^{-1}$ )	$^3\text{He}/^4\text{He}^*$ ( $R/R_a$ )	$^4\text{He}^\dagger$ ( $10^{-9}\text{cm}^3\text{STP g}^{-1}$ )	$^3\text{He}/^4\text{He}^\dagger$ ( $R/R_a$ )	$^{143}\text{Nd}/^{144}\text{Nd}^\ddagger$	$^{87}\text{Sr}/^{86}\text{Sr}^\ddagger$	(La/Sm) <sub>n</sub>	ΔNb
Cape Searle								
CS/19	10.0 ± 0.3	23.0 ± 1.3			0.512924 ± 6	0.703420 ± 17	1.82	0.26
CS/7	130.6 ± 0.6	43.9 ± 0.6	33.6 ± 0.17	7.4 ± 0.5	0.512730 ± 11	0.704912 ± 17	0.92	-0.11
CS/6	20.9 ± 0.2	37.9 ± 1.2	6.9 ± 0.58	4.7 ± 0.7	0.512929 ± 7	0.703445 ± 20	1.78	0.20
CS/5	4.4 ± 0.2	26.3 ± 2.0	169.6 ± 0.86	0.16 ± 0.04	0.512921 ± 13	0.703314 ± 20	1.86	0.19
Padloping Island								
PI/23	39.8 ± 0.2	43.9 ± 0.6	23.4 ± 0.12	2.1 ± 0.1	0.513051 ± 8	0.703248 ± 20	1.00	-0.17
PI/25	11.0 ± 0.1	49.5 ± 1.6	164.7 ± 0.82	0.12 ± 0.02	0.513030 ± 6	0.703142 ± 24	1.04	-0.05
PI/26	24.1 ± 0.1	43.9 ± 0.5	6.6 ± 0.05	17.3 ± 0.8	0.513019 ± 6	0.703181 ± 24	1.15	-0.23
PI/27	45.6 ± 0.2	43.1 ± 0.5	11.8 ± 0.07	9.9 ± 1.2	0.513026 ± 8	0.703076 ± 21	1.00	-0.12
Durban Island								
DI/23	22.4 ± 0.3	14.5 ± 0.5	64.3 ± 0.33	2.1 ± 0.13	0.512845 ± 9	0.703964 ± 20	2.25	0.26
DI/24	5.1 ± 0.04	37.5 ± 0.6	6.2 ± 0.33	10.5 ± 0.8	0.513012 ± 6	0.703572 ± 20	1.22	-0.25
DI/27	17.4 ± 0.4	38.6 ± 1.5	7.0 ± 0.08	6.9 ± 0.8	0.512976 ± 8	0.703372 ± 21	0.93	-0.23

\*Helium was extracted by *in vacuo* crushing 1–2 g of olivine and analysed at SUERC using procedures reported previously<sup>19</sup>.  $^3\text{He}/^4\text{He}$  ratios ( $R$ ) are normalized to the atmospheric ratio ( $1.39 \times 10^{-6}$ ;  $R_a$ ) and corrected for blanks.  $^4\text{He}$  blanks averaged  $4 \times 10^{-11}\text{cm}^3\text{STP}$  and  $^3\text{He}$  blanks ( $5 \times 10^{-15}\text{cm}^3\text{STP}$ ) were always less than 5% of the measured  $^3\text{He}$ . Correction for atmospheric He on the basis of Ne abundances is insignificant.

†Olivine powders (~100 mg) produced by *in vacuo* crushing were melted at 1,850 °C in a double-walled resistance furnace. Hot furnace  $^4\text{He}$  blanks averaged  $2 \times 10^{-11}\text{cm}^3\text{STP g}^{-1}$ .  $^3\text{He}$  blanks were the same as crush extractions.

‡Whole-rock Sr and Nd isotopic ratios<sup>28</sup> have been corrected to 60 Myr ago on the basis of measured Rb/Sr and Sm/Nd. This reduces  $^{87}\text{Sr}/^{86}\text{Sr}$  by less than 0.007% and  $^{143}\text{Nd}/^{144}\text{Nd}$  by less than 0.002%. Uncertainties (2σ) are in the last decimal place. Rare-earth element concentrations were measured by ICP-MS<sup>29</sup>. (La/Sm)<sub>n</sub> is chondrite normalized, and has an uncertainty of ± 3% (2σ). Nb, Y and Zr concentrations were measured in triplicate by X-ray fluorescence and the averages used<sup>24</sup>. ΔNb =  $1.74 + \log(\text{Nb}/\text{Y}) - 1.92 \times \log(\text{Zr}/\text{Y})$ <sup>30</sup>. Uncertainty in ΔNb never exceeds ± 10%.

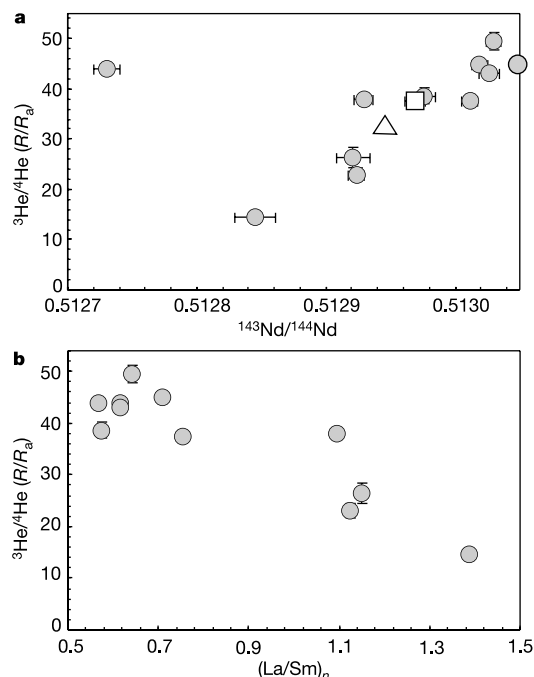
$^3\text{He}/^4\text{He}$ . Pb isotopes are sensitive to crustal contamination in North Atlantic margin Palaeocene basalts, because the Precambrian basement has distinctively unradiogenic Pb (ref. 18). The Baffin Island picrites show no correlation between Pb isotope composition and other isotopic tracers (Sr, Nd, He). Thus, while the Pb isotopes may indicate some crustal interaction, its effect on other elements appears negligible. We have previously demonstrated that basalts whose whole-rock Pb isotope compositions are clearly affected by crustal contamination retain uncontaminated He (ref. 19), either because the olivine crystallized before crustal assimilation, or because the deep crust is He-poor. In this study, we observe a similar effect: dyke sample CS/7 has high  $^3\text{He}/^4\text{He}$  ( $43.3 \pm 0.6R_a$ ) but has high  $^{87}\text{Sr}/^{86}\text{Sr}$  and low  $^{143}\text{Nd}/^{144}\text{Nd}$  that are best attributed to crustal contamination. As crustal contamination is expected to lower  $^3\text{He}/^4\text{He}$ , we conclude that crush-release He is insensitive to any contamination. The inability of magmatic or post-magmatic processes to alter He isotope ratios, and the strong correlation with Sr and Nd isotopes and trace elements (see below), means that the sample (PI/25) with the highest helium isotope ratio provides the best estimate yet ( $49.5 \pm 1.5R_a$ ) of the minimum He isotope composition of the less degassed mantle reservoir.

The olivine  $^3\text{He}/^4\text{He}$  ratio correlates strongly with whole-rock  $^{143}\text{Nd}/^{144}\text{Nd}$ ,  $^{87}\text{Sr}/^{86}\text{Sr}$  and trace-element ratios such as La/Sm (see Fig. 1). The absence of co-variation of  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  with indices of contamination such as Ba/Nb and Pb isotope ratios suggests that crustal contamination has not significantly affected Sr and Nd isotopes (with the exception of the dyke sample CS/7). Furthermore, high- $^3\text{He}/^4\text{He}$  Palaeocene basalts from Iceland<sup>3</sup> that cannot have been contaminated by continental crust have He–Nd and He–Sr isotopic compositions that plot on the trend. Consequently, the He isotope correlations with La/Sm and the lithophile isotope ratios record binary mixing between large-volume melts of a  $^3\text{He}$ -rich, relatively depleted mantle reservoir, and small-volume melts from a mantle reservoir with low  $^3\text{He}/^4\text{He}$ , rich in incompatible elements.

The depleted mantle end-member inferred from the most extreme Sr and Nd isotope compositions requires a mantle source region with  $^{87}\text{Sr}/^{86}\text{Sr} \approx 0.70305$ ,  $^{143}\text{Nd}/^{144}\text{Nd} \approx 0.51305$  and  $^3\text{He}/^4\text{He} > 50R_a$ . The Sr and Nd isotope ratios are indistinguishable from the most depleted basalts erupted on Iceland and only slightly less depleted than North Atlantic MORB erupted away from the Iceland plume ( $^{87}\text{Sr}/^{86}\text{Sr} < 0.7030$ ,  $^{143}\text{Nd}/^{144}\text{Nd} > 0.5131$ ). Icelandic and North Atlantic MORBS define parallel, non-overlapping arrays on a plot of  $\log(\text{Nb}/\text{Y})$  against  $\log(\text{Zr}/\text{Y})$ . The excess or

deficiency in Nb is defined by the ΔNb notation such that Icelandic basalts have ΔNb > 0 and MORB have ΔNb < 0 (ref. 24). The high  $^3\text{He}/^4\text{He}$  Baffin Island basalts have strongly negative ΔNb (Table 1) indicating derivation from the North Atlantic MORB-source mantle rather than the depleted Iceland plume.

It is, first, important to note that this He-, Sr- and Nd-isotopic composition cannot represent MORB source mantle at 61 Myr ago as the  $^3\text{He}/(\text{U}+\text{Th})$  of the MORB reservoir is incapable of significantly changing  $^3\text{He}/^4\text{He}$  in hundreds of millions of years. If He is less incompatible than U during melting, high- $^3\text{He}/^4\text{He}$  would correspond with the most depleted basalts<sup>20</sup>. Low- $^{87}\text{Sr}/^{86}\text{Sr}$ , high- $^{143}\text{Nd}/^{144}\text{Nd}$  and high- $^3\text{He}/^4\text{He}$  basalts are indicative of a



**Figure 1** Plots of olivine  $^3\text{He}/^4\text{He}$  versus  $^{143}\text{Nd}/^{144}\text{Nd}$  and (La/Sm)<sub>n</sub> for early Palaeocene basalts from Baffin Island. **a**, Whole-rock Nd isotopic composition; also shown are the data for a Palaeogene ankaramite from NW Iceland (square) and basaltic glass from Loihi seamount (triangle). **b**, Whole-rock (La/Sm)<sub>n</sub>.

depleted, 'regassed' component which would be generated in a reservoir with low U/He which grows  $^4\text{He}$  less quickly than MORB source mantle. The mechanism proposed to generate the low-U/He source<sup>20</sup> requires (shallow) melt-gas separation to fractionate He from U and the other lithophile trace elements. It is difficult to reconcile the linear He–Sr and He–Nd isotope arrays (Fig. 1) with such a component since a high-He/Sr and -He/Nd ratio component mixed with 'normal' He/Sr and He/Nd mantle would result in strongly hyperbolic mixing arrays. Instead, we make the assumption that the dominant control on isotopic heterogeneity in the mantle is the same for all elements—that is, solid-melt distribution coefficients—and that He is highly incompatible during mantle melting<sup>21</sup>. Therefore any reservoir with low Rb/Sr and high Sm/Nd, because it is residual to partial melting, will have lost most of its He and would evolve low- $^3\text{He}/^4\text{He}$ , low- $^{87}\text{Sr}/^{86}\text{Sr}$  and high- $^{143}\text{Nd}/^{144}\text{Nd}$  ratios. The subduction of interplanetary dust particles is incapable of delivering enough solar noble gases to the upper mantle to change its He and Ne isotopic composition significantly<sup>22</sup>. In the absence of alternatives we show below that the high- $^3\text{He}/^4\text{He}$  component is most probably the product of mantle mixing.

If Earth accreted with  $^3\text{He}/^4\text{He} = 120R_a$ , bulk silicate Earth U (21 p.p.b.) and U/Th = 3.8, an undegassed mantle reservoir with  $^3\text{He}/^4\text{He} = 50R_a$  has a  $^3\text{He}$  concentration of  $8 \times 10^{10}$  atoms  $\text{g}^{-1}$ . If moderately incompatible lithophile element concentrations in the depleted mantle reservoir are less than a factor of two lower than undepleted mantle<sup>23</sup>, mixing a small proportion of  $^3\text{He}$ -rich mantle with depleted upper mantle ( $1.2\text{--}4.6 \times 10^9$   $^3\text{He}$  atoms  $\text{g}^{-1}$ ; ref. 6) severely changes  $^3\text{He}/^4\text{He}$  without significantly altering trace element concentrations and Sr and Nd isotope ratios. Consequently the isotopic composition of the high  $^3\text{He}/^4\text{He}$  end-member can be generated by mixing depleted mantle and primordial  $^3\text{He}$ -rich mantle reservoir. The Baffin Island data define approximately linear mixing arrays in He–Sr and He–Nd isotope space, implying that the He/Nd and He/Sr ratios and, therefore, He concentrations, of both reservoirs are similar. The absence of strongly hyperbolic mixing trends supports the contention that the high  $^3\text{He}/^4\text{He}$  end-member is not simply undegassed mantle.

The enriched mantle end-member, estimated by extrapolating the He–Nd and He–Sr isotope trends to the most extreme values measured in uncontaminated Baffin Island basalts ( $^{143}\text{Nd}/^{144}\text{Nd} = 0.51282$ ;  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7042$ ), has a  $^3\text{He}/^4\text{He}$  similar to, or lower than, the MORB ratio ( $8 \pm 1 R_a$ ). The positive Nb value of this end-member, and the coincidence of the high  $^3\text{He}/^4\text{He}$  ankaramite from northwest Iceland<sup>3</sup> with the mixing trend, supports an origin for this enrichment in the mantle asthenosphere. Although the ultimate origin is unclear, the low  $^3\text{He}/^4\text{He}$  ratio requires that it has been isolated from the high- $^3\text{He}/^4\text{He}$  reservoir prior to melting. It may represent fusible heterogeneities in the upper mantle or the plume that are large enough to prevent isotopic homogenization yet small enough to be preferentially sampled by melting.

The convergence of Sr–Nd–Pb isotope trends in ocean island basalt (OIB) and MORB have been used to identify a common deep mantle reservoir that is the carrier of the high- $^3\text{He}/^4\text{He}$  plume component. The most primordial  $^3\text{He}$ -rich basalts from Iceland and Hawaii lie on the He–Nd and He–Sr isotope mixing trends identified here suggesting that the extreme end of the Baffin Island picrite trend is the best estimate of a common, high- $^3\text{He}/^4\text{He}$  mantle component. This is at the depleted end of the ranges proposed previously for a common mantle reservoir<sup>24,25</sup>, but the negative  $\Delta\text{Nb}$  requires that it originates in the depleted upper mantle, rather than the lower mantle. The thermal boundary layer between upper- and lower-mantle regions is one potential reservoir for such material<sup>26,27</sup>, although it cannot be ruled out that the mixing occurs as the plume head disperses in the upper mantle. The dominance of the  $^3\text{He}$ -recharged, depleted component in mantle plumes is consistent with a low mass flux from the lower mantle and explains why

basalts from Iceland and Hawaii appear to have lower He concentrations than predicted for undegassed mantle<sup>9,10</sup>. □

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